Nonlinear lattice dynamics in two dimensions

Daniel C. Mattis
Physics Department, University of Utah, Salt Lake City, Utah 84112
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In general, dynamical and quantum-mechanical properties of anharmonic lattices are best expressed in terms of bond variables. A transformation from atomic (site) to bond coordinates is generated in two dimensions, with the help of a number of subsidiary conditions expressed on plaquettes. It is shown that these commute with the Hamiltonian and yield a number of zero-energy modes. With the use of this trick, two-dimensional problems are now only slightly more difficult to study than the one-dimensional analogs. We discuss some exact results.

Because the elastic energy of a lattice is stored in its interatomic bonds, it would be convenient to be able to express all dynamic variables in terms of bond coordinates. The transformation from site-centered coordinates (e.g., displacement of individual atoms from their ideal lattice positions) to bond coordinates (separation of nearest-neighbor atoms) is straightforward in one dimension (1D) but is rather tricky in 2D. In the present work we devise a transformation, not unlike the duality transformation of José, Kadanoff, Kirkpatrick, and Nelson,[1] which permits us to attain this goal in 2D. We can then formulate models of anharmonic interactions and solve them with practically the same ease as in 1D.[2]

Long ago, Peierls[3] noted that the thermal excitation of long-wavelength phonons is sufficient to destroy long-range crystalline order in 2D, an observation reinforced by the rigorous theorem of Mermin and Wagner.[4] If the deviations of each atom from its "ideal" position are large, then lattice dynamics *cannot* be based on the usual assumption on which the usual theories of phonons are based—that of small deviations from ideal positions. On the other hand, deviations of nearest-neighbor atom *separations* from their ideal values can be relatively small, even in 1D or 2D, and even at high temperatures. Thus anharmonic approximation in terms of bond coordinates can make sense even when the usual theory does not.

In the Kosterlitz-Thouless[5] and Nelson-Halperin[6] theories of two-dimensional melting, the role of dislocations is crucial. It would be interesting to see whether a model free of dislocations, but in which the anharmonicity of the bonds is taken into account, is capable of reproducing some of the features which are now believed to be associated with two-dimensional melting. The present paper provides the theoretical and mathematical framework in which to do this.

The transformation from site to bond coordinates is simple only in 1D.[2] There we define the bond coordinate r_n in terms of the site coordinates x_n as $r_n = x_n - x_{n-1}$. If $\hat{p}_n$ is the momentum conjugate to x_n, then $p_n - p_{n+1} = \hat{p}_n$, with p_n conjugate to r_n. The one-dimensional Hamiltonian

$$H = \frac{1}{2M} \sum \hat{p}_n^2 + \sum V(x_n - x_{n-1}) \ ,$$

originally written in terms of site coordinates, becomes

$$H = \frac{1}{M} \sum (p_n^2 - p_n p_{n+1}) + \sum V(r_n) \ .$$

The advantages of this transformation were made clear in Ref. 2. At present, we demonstrate the transformation to bond coordinates in 2D, then construct a number of models in this representation and solve two of them completely.

Before proceeding, it is important to show why the simple transformation of Ref. 2 is inoperative. If we measure each site from its left-hand nearest neighbor, the vertical nearest neighbors will be quite distant in the numbering scheme. For example, indicating the sites on one row by x_n and those on the neighboring row by x_n', the nth *vertical* bond connecting x_n and x_n' would take the form

$$V\left(\sum_{m=0}^{n} (r_m' - r_m) \right) \ .$$

Thus, violation of the symmetry of the lattice (rotation by 90° interchanges vertical and horizontal bonds) leads to ugly representation of the vertical bonds, even though the horizontal bonds have the nice form, $V(r_n)$ or $V(r_n')$.

The remedy is simply to *take* the bonds as our new dynamical variables, then to introduce appropriate compatibility equations. Happily, these are satisfied trivially. Consider first a single square of four atoms, as in Fig. 1. The individual atoms have one, two, or three degrees of freedom according to the model we wish to consider. Let us restrict them to one degree

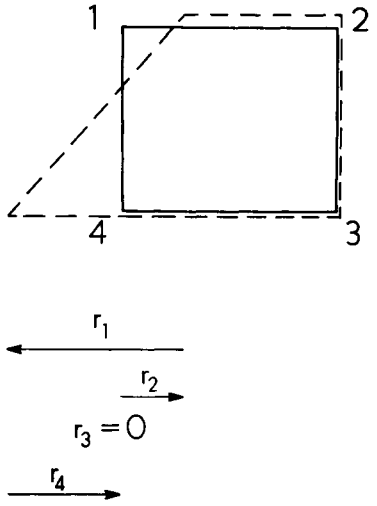

FIG. 1. Four sites form an elementary plaquette (solid lines). After displacement, the bonds (dashed lines) assume deviations $r_1, \ldots, r_4$, subject to constraint Eq. (1). In this figure we only show displacements parallel to the x axis.

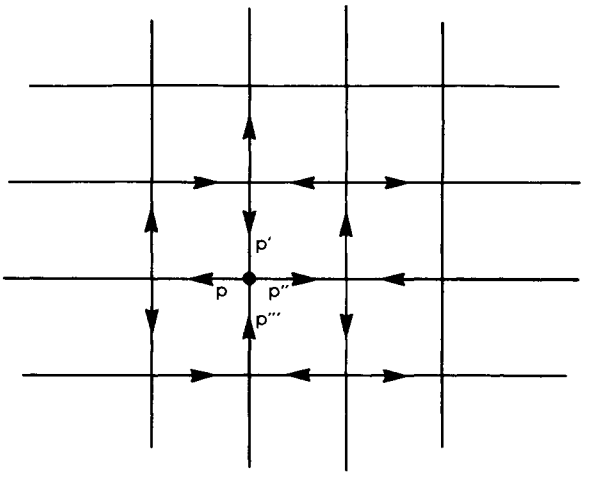

FIG. 2. Assignment of bond orientations in a two-dimensional square lattice. The constraint (1) applies to each plaquette individually. The momenta at each vertex add according to the sense of the arrows, yielding a kinetic energy for each vertex: $(1/2M)(p+p''-p'-p''')^2$. When decomposed into bond-diagonal (e.g., p^2) and bond-linking (e.g., pp'') terms, this leads to the bond-diagonal H_n^0 of Eq. (4) and the H'_{nm} bond-bond couplings in Eq. (5).

of freedom here, motion in the x direction. The distorted square satisfies

$$r_1+r_2+r_3+r_4=0 \ , \tag{1}$$

where $r_n=x_n-x_{n-1}$ (with $r_1=x_1-x_4$). In bond variables, the Hamiltonian takes the form

$$H=\frac{1}{2M}[(p_2-p_1)^2+(p_3-p_2)^2+(p_4-p_3)^2 + (p_1-p_4)^2]+\sum_{n=1}^{4}V(r_n) \ . \tag{2}$$

We now note that the quantity in (1) commutes with the operator (2), therefore condition (1) can always be satisfied. The example of harmonic oscillators, $V(r)=\frac{1}{2}Kr^2$, is instructive. Equation (2) is easily diagonalized for this case, and it is seen that only the $q=0$ phonons involve the quantity in (1). But by an obvious symmetry, these phonons have *zero* energy. It is therefore trivial to construct a state satisfying (1), $\delta(r_1+r_2+r_3+r_4)$, which carries no energy, and multiplies all eigenstates $\phi(r_1,r_2,\ldots,r_4)$ of (2).

Now consider for the moment the full two-dimensional lattice, again restricting motion to the x direction. Taking each nearest-neighbor bond provides us with $2N$ variables. We now impose a condition (1) on *each* plaquette. This provides N linear relations among our $2N$ variables, restoring the correct number of dynamical variables. The question will remain—how to satisfy these linear relations; and we shall see that this occurs as trivially as for the single square, if we order the sense of the bonds according to the system of arrows given in Fig. 2.

The sense of an arrow is given by the definition $r_n=x_n-x_{n-1}$; the arrow starts at n and points at $n-1$. With the arrows distributed as in Fig. 2 (or any trivial translation thereof) the sum rule becomes *exactly Eq. (1) for each plaquette.* In terms of canonical bond momenta p_n, the original atomic momentum $\hat{p}_j$ for the jth site becomes $\hat{p}_j=p_1+p_2-p_3-p_4$, where bonds 1,2,3,4 all emanate from this site. The sign $\pm$ is chosen according to the sense of the arrow. If the arrow is directed away from j, choose $+$. If the arrow points into the site j, choose $-$.

Each site is surrounded by four plaquettes, and the momentum operator at the site contains two p's associated with each of these plaquettes. Because of the sense of the arrows, these p's always come in with opposite signs, and therefore the site momentum commutes with the plaquette conditions (1). It follows that H commutes with the set of N conditions (1), so that these will be trivially satisfied by zero-energy modes just as in the case of the single square.

Finally, H takes the convenient form

$$H=\sum_n H_n^0+\sum_{n,m}H'_{nm} \ , \tag{3}$$

where each bond Hamiltonian is

$$H_n^0=\frac{1}{M}p_n^2+V(r_n) \ , \tag{4}$$

and the interaction parts are

$$H'_{nm}=\frac{1}{M}p_np_m\epsilon_{nm} \ , \tag{5}$$

where n,m are two contiguous bonds and $\epsilon_{nm}\equiv+1$ if

their arrows both point into (or out of) the same vertex, and $\epsilon_{nm} \equiv -1$ otherwise. With our assignment of arrows, Fig. 2, this is equivalent to the statement: $\epsilon_{nm} = +1$ if both contiguous bonds are horizontal or vertical, and $\epsilon_{nm} = -1$ if one is vertical and the other horizontal.

It may be redundant to repeat that H is augmented by the set of N subsidiary conditions (1) about each plaquette, but this *is* an important feature.

So far we have considered only motions along the x direction. If, additionally, we now permit motion along the y or z directions, *all* our arguments go through *mutatis mutandis;* dynamical variables in (1), (4), and (5) are now to be treated as vectors, and scalar multiplication taken where appropriate $(\vec{p}_n \cdot \vec{p}_m)$. Although the lattice is two dimensional, in some cases it may be appropriate to allow each atom (or bond) three-dimensional motion; whereas in other cases, there may be additional constraints on the vertical motion, and two-dimensional motion may be all we need consider.

Turning to applications let us begin with *harmonic oscillators.* The perfect harmonic lattice, $V(r) = \frac{1}{2}Kr^2$, can be solved in the original coordinates to reveal the usual phonon spectrum. In our new bond language, we recover precisely the same spectrum plus N (or $2N$ or $3N$ depending on whether motion is allowed along 1, 2, or 3 orthogonal directions) zero-energy modes. These have no effect on the dynamics but would cause the partition function to diverge if we blindly included them in the trace. The N (or $2N$ or $3N$) subsidiary conditions (1) ensure that each such mode factors precisely 1 into the partition function, and thus has no more thermodynamic than it had dynamical consequences. Some details are supplied in the appendix.

A second model, introduced in our study of the quantum-mechanical molecule, considers the case when (4) has effectively *only two levels.* In terms of Pauli matrices, the Hamiltonian takes the form of an unusually bonded (highly frustrated) two-dimensional Ising model in a transverse magnetic field:

$$H_n^0 = J_0 \sigma_n^z \tag{6}$$

and

$$H'_{nm} = J_1 \sigma_n^x \sigma_m^x \epsilon_{nm} \ . \tag{7}$$

Because of the ϵ_{nm}, (7) does not represent purely ferromagnetic or antiferromagnetic coupling. Thus, unlike the analogous model in one dimension,[2] it is not obvious that the ground state has a discontinuity at a critical J_1/J_0, nor is it obvious that the free energy has a discontinuity (phase transition) at some finite T_c. The Hamiltonian (6) and (7) must be augmented by the N subsidiary conditions (1), proportional to the unit matrix (since they necessarily commute with H). In calculating the partition function associated with this Hamiltonian, it may be important to factor out the plaquette Goldstone bosons, i.e., the redundant zero-energy configurations. (The precise procedure is not obvious at the present time, so it is possible that the above H is a nontrivial generalization of the one-dimensional model treated in Ref. 2.)

We now turn to a third, anharmonic model considered in Ref. 2. Here the atoms are *loosely coupled,* and the lattice "melts" spatially at $T = 0$. The bonds between two horizontally neighboring atoms are constrained such that $(a - \frac{1}{2}d) < x_n - x_{n-1} < (a + \frac{1}{2}d)$, $-\frac{1}{2}d < y_n - y_{n-1} < +\frac{1}{2}d$, and $-\frac{1}{2}d < z_n - z_{n-1} < +\frac{1}{2}d$. Boundary conditions such as

$$\phi_{y_n - y_{n-1} = 1/2d} = \phi_{y_n - y_{n-1} = -1/2d} \tag{8}$$

are imposed, so that within each well plane-wave solutions are obtained. With $\vec{k}_n = (k_n^x, k_n^y, k_n^z)$ and each $k_n^i = 2\pi x N/d$ (N an integer), the total energies are

$$E = \frac{\hbar^2}{M}\left(\sum_n \vec{k}_n^2 + \sum_{n,m} \vec{k}_n \cdot \vec{k}_m \epsilon_{nm} \right) \ . \tag{9}$$

For this model we can calculate the partition function. First we note that the energies (9) can be written in a form involving only the vertices:

$$E = \frac{\hbar^2}{2M}\left(\sum_n (\vec{k}_{n,1} + \vec{k}_{n,2} - \vec{k}_{n,3} - \vec{k}_{n,4})^2 \right) \ , \tag{10}$$

where $\vec{k}_{n,i}$ are the four $\vec{k}$'s connecting into each vertex and the sign (± 1) is taken according to the direction of the arrow into the vertex. Now it is obvious that if momentum is conserved at a vertex (the sum of the k's vanishes) its contribution to the energy vanishes. Constructing the two-dimensional lattice by adding row upon row, we see that whenever we add a vertical bond to a vertex, its value can be chosen to make the energy of that vertex equal to any value of

$$\frac{\hbar^2}{2M}(n_x^2 + n_y^2 + n_z^2)(2\pi/d)^2 \ ,$$

where n_x, n_y, n_z, are any three integers (for three-dimensional motion; only n_x and n_y for two-dimensional motion, and only n_x for one-dimensional motion). The two horizontal bonds that attach to the new vertical bond are irrelevant to the energy and therefore represent the zero-energy modes. Eliminating these, we obtain, for the free energy per site,

$$F/N = -kTq \ln\left[\sum_{n=-\infty}^{+\infty} \exp - \frac{\hbar^2}{2MkT}\left(\frac{2\pi n}{d}\right)^2 \right] \ , \tag{11}$$

in which $q = 1, 2, 3$, for one-, two-, and three-dimensional motion.

This result, reminiscent of what we obtained for this loose lattice in 1D, shows there is no phase transition for this model of $V(r)$. It should be noted

that this "loose model" shares a defect with the harmonic model, viz., $V(r)$ has *only bound states.* Any *reasonable* model of $V(r)$ allows for a finite number (perhaps several dozen) bound states, and also an infinite number of continuum (ionized) states. However closely the bound states may conform to the harmonic-oscillator picture, the continuum states certainly do not. Therefore, at finite temperature, a finite number of bonds must be "broken," resulting in vacancies, interstitials, and even dislocations. It would seem that the proper choice of $V(r)$ would lead to a first-principles theory of a two-dimensional solid, including defects, tractable because of the decompositions (3)–(5).

Our work will continue in that direction, along with our procedure to generalize the analysis to 3D.[7]

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APPENDIX

The unit cell of the bond lattice contains two bonds as its basis: a vertical (a) and a horizontal (b) bond. We Fourier transform H. The kinetic energy assumes the form

$$\frac{2}{M}\sum_k |p_{ka}\sin\tfrac{1}{2}k_y + p_{kb}\sin\tfrac{1}{2}k_x|^2 \ , \tag{A1}$$

whereas the potential energy, for harmonic-oscillator potentials, becomes

$$\tfrac{1}{2}K\sum_k (|r_{ka}|^2+|r_{kb}|^2) \ . \tag{A2}$$

It is simplest to look at the equations of motion. We observe

$$-\frac{d^2}{dt^2}(r_{ka}\sin\tfrac{1}{2}k_y + r_{kb}\sin\tfrac{1}{2}k_x) = \Omega_k^2(r_{ka}\sin\tfrac{1}{2}k_y + r_{kb}\sin\tfrac{1}{2}k_x) \ ,$$

with

$$\Omega_k \propto (\sin^2\tfrac{1}{2}k_x + \sin^2\tfrac{1}{2}k_y)^{1/2} \ , \tag{A3}$$

using the usual commutator bracket equations of motion. This result reproduces the known dispersion for the simple square lattice of harmonic oscillators. On the other hand,

$$\frac{d}{dt}(r_{ka}\sin\tfrac{1}{2}k_x - r_{kb}\sin\tfrac{1}{2}k_y) = 0 \tag{A4}$$

supplies us with N constants of motion. In a quantum-mechanical treatment, we would factor out the states relating to the dynamical degrees of freedom in (A4). In classical dynamics, we would make sure the N constraints corresponding to Eq. (1) of the text are satisfied at $t=0$; the equations of motion (A4) would then guarantee their satisfaction at all t.

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[7]After this present paper was submitted, we discovered a procedure for transforming to bond variables in arbitrary lattices including all three-dimensional lattices, amorphous lattices, and in higher dimensions. However, because the procedure outlined in the present paper is optimal for the square lattice, it was not deemed necessary to rewrite the present work. Details concerning the generalizations will be published [D. Mattis, Phys. Lett. (in press)].